

A52D-08 1635h

Advances in Measurement of Carbonyls in Aerosols.

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Chamber studies establish the formation of highly polar oxygenated species from the reaction of anthropogenic and biogenic hydrocarbons with hydroxyl radicals or ozone. A paucity of data exists however on the generation and fate of these organics in the ambient atmospheric environment. This is primarily due to the absence of suitable analytical methods. To address limitations of existing methods, we developed methods that rely on O-(2,3,4,5,6)-pentafluorobenzylhydroxylamine (PFBHA), and bis(trimethylsilyl) trifluoroacetamide (BSTFA) in concert with GC/ion trap mass spectrometry (GC/ITMS) to identify and quantify carbonyl, dicarbonyl and hydroxy carbonyl photooxidation products in aerosols at part-per-trillion (pptv) levels. We also optimized and evaluated a mist chamber to sample carbonyls and multi-functional carbonyls with 10 minute sampling times. We applied the method to identify and quantify 2-hydroxy-2-methyl propanal (2-HMPR), a proposed photooxidation product of 2-methyl-3-buten-2-ol (MBO) in the Blodgett Forest, CA. The average 2-HMPR/MBO mixing ratio was 0.33 0.25, which is reasonable since the expected yield of 2-HMPR from the hydroxyl radical oxidation of MBO is 0.19-0.35. Further method development in our laboratory is exploring the employment of HPLC/atmospheric pressure chemical ionization (APCI) mass spectra to identify model aliphatic and aromatic carbonyls (the major classes were aldehydes, ketones, dicarbonyls, and quinones) in aerosols. The data indicate the potential for pentafluorobenzyl derivatization in concert with GC/ITMS and HPLC/ITMS to measure a broad range of carbonyls.

A52E MCC: 102 Friday 1330h

Transport and Effects of Anthropogenic Pollutants: ITCT

2K2, Including PEACE I (joint with GC)

Presiding: Y Kondo, The University of Tokyo; D Parrish, NOAA Aeronomy Laboratory

A52E-01 1330h INVITED

Chemical air mass characteristics along the West Coast of the United States during April 2002, Results from ITCT 2002

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In April-May of 2002 the concentration of ozone, ozone precursors, and other photochemical products were measured during 13 flights on board of the NOAA WP3 aircraft over the Eastern Pacific Ocean and the Western United States as part of the Intercontinental Transport and Chemical Transformation (ITCT) experiment. The experiment characterized the long range transport across the Pacific and the addition of fresh emissions along the west coast of the United States.

Extensive pollution layers were observed on several of the flights and allow the study of the chemical transformations during distinct transport regimes.

A52E-02 1350h INVITED

Impact of Asian Outflow on O3 and its Precursors Over Japan in January 2002

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The Pacific Exploration of Asian Continental Emission Phase A (PEACE-A) aircraft measurement campaign was conducted over Japan in January 2002. In situ aircraft measurements of ozone (O₃), carbon monoxide (CO), reactive nitrogen (NO and NO_y), carbon dioxide (CO₂) non-methane hydrocarbons (NMHCs), and other species were made during PEACE-A. The flights were conducted over the Japan Sea (35-45 N) and East China Sea (20-35 N), providing useful data set to investigate the horizontal and vertical distributions of trace gases in these regions during wintertime. Large enhancements of CO, CO₂, NO_y, and NMHCs were frequently observed in the boundary layer (< 4 km), while the mixing ratios of these species in the free troposphere were fairly constant. The origins of air masses sampled in the boundary layer are roughly estimated using backward trajectories. In addition, we use dCO/dCO₂ ratios in the observed air masses (linear regression slope of CO-CO₂ correlation) as a diagnostics of combustion types of emission sources. The air masses sampled at 0-2 km over the Japan Sea were mostly transported from northern China and dominated by relatively high combustion efficiency sources (dCO/dCO₂ = 0.02-0.04). By contrast, the air masses sampled at 2-4 km over the East China Sea were mostly transported from southern China and significantly affected by lower combustion efficiency sources (dCO/dCO₂ ratios = 0.04-0.15). Correlations among CO, NO_x, NO_y, and O₃ are discussed in terms of the difference in the dCO/dCO₂ ratios to evaluate the contribution from each type of emission source to the regional budgets of reactive nitrogen and O₃ during the PEACE-A period.

A52E-03 1410h INVITED

Forecasting and Modeling Trans-Pacific Transport of Asian Pollution in ITCT2K2

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We used the GEOS-CHEM global 3-D model of tropospheric chemistry driven by forecast meteorological fields from the Goddard Earth Observing System (GEOS) to forecast transpacific transport events

during the ITCT2K2 aircraft mission over the west coast in April-May 2001. ITCT2K2 focused on characterizing Asian inflow to North America. The forecast simulations transported five tagged CO tracers from different source regions. The model successfully predicted several transpacific transport events that were confirmed by in situ observations. We further investigate the performance of the model forecasts by comparing forecast results to the observations and to the post-mission full-chemistry simulations driven by re-analysis meteorological fields. The model is evaluated with the in situ aircraft and ground measurements from both ITCT2K2 and the PEACE-B aircraft mission. The chemical evolution of Asian pollution during transport and the seasonal variations of transpacific transport are investigated.

A52E-04 1430h INVITED

Measurements at Trinidad Head, California during ITCT 2K2: Were Asian emissions observed?

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Measurements for a wide suite of trace gases and aerosols were made at Trinidad Head, California, from 19 April through 22 May 2002 as part of the NOAA ITCT research program. This talk will provide an introduction to the ITCT ground site, and will address one of the major scientific questions for the measurement campaign: What is the influence of Asian emissions on air masses entering North America in spring-time? CO has been identified as one of the most useful tracers to look for Asian emission plumes because of its relatively long atmospheric lifetime and its emission from all combustion sources. Before assessing the influence of long range transport on the observations, local/regional influences must be filtered out of the data set. Methyl Tertiary Butyl Ether (MTBE) serves as a useful tracer because its emissions are associated with use as a fossil fuel additive in North America and its atmospheric lifetime is a few days. CO₂ and radon also serve as useful tracers of regional continental influences, particularly at night when their concentrations are enhanced due to emissions under stable atmospheric conditions. Filtering out local influences removed 25 to 50% of the observations, depending on the constraints applied, and it decreased the mean CO mixing ratio by 4% to 147 ppb. After filtering the data to remove local influences, the variability and the absolute concentration in the remaining CO data can be examined for Asian influence by comparison with forecast models run as part of the ITCT campaign such as GEOS-CHEM (Harvard), MOZART (NCAR), and CFORS (IOWA). The observed variability in the filtered CO data is in general well simulated by the models, showing a dominant influence from North American fossil fuel emissions. Distinct Asian pollution plumes could not be identified in these ground based observations, because the magnitude of CO variability due to Asian emissions was small relative to the total observed variability. However, the relative contributions of emission sources to the total observed CO could be approximated through comparison of the measurements and model results. Comparison of all the ground based trace gas and aerosol measurements with the model results should utilize similar techniques to remove local influences that are unlikely to be captured by the models. Filtering out local influences decreases mean concentrations of aerosols and ozone precursors such as volatile organic compounds and oxides of nitrogen, but it increases the mean concentration of ozone.

A52E-05 1510h

Airborne Observations of PAN and Related Compounds During ITCT-2K2

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Peroxyacetic Nitric Anhydride (PAN), and related homologues, were measured aboard the NOAA WP3 aircraft during the ITCT-2K2 experiment, as part of an extensive set of chemical measurements. The data show that PAN is often the most abundant odd-nitrogen (NOy) species in the Eastern Pacific, and the median PAN/NOy is 0.5 at 5 to 8 km altitude. Several interesting transport events were observed where outflow of pollutants from Asia was intercepted just off the coast of North America. There was also evidence for layers at 35 km altitude that had been depleted in PAN, possibly because of processing of air masses through the marine boundary layer (MBL). The importance of PAN as a means of intercontinental transport of NOy will be discussed in relation to these measurements.

A52E-06 1525h

Observations of organic trace gases during ITCT: Characterization of sources, background, and long-range transport to the US West coast and eastern Pacific atmosphere

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The NOAA ITCT (Intercontinental Transport and Chemical Transformation) mission examined the processes that impact the chemical composition of the atmosphere in the eastern North Pacific Ocean and along the West Coast of the United States. The mission took place during April - May, 2002, when long-range transport to the US West Coast from downwind sources in Asia and beyond is most favorable. As part of the atmospheric chemistry payload on the NOAA P3 aircraft, whole air samples were collected for analysis of a variety of organic trace gases, including methane, NMHC, halocarbons, organic nitrates, and selected sulfur species. Mission flight tracks were designed to examine regions characteristic of the background atmosphere, and regions impacted by specific point sources, larger urban sources, and long-range transport. In this

presentation we summarize the organic trace gas measurements and relationships from the whole air samples, characterize signatures of emissions from the west coast urban areas, and identify signatures of long-range trans-Pacific transport.

A52E-07 1540h

VOC Composition of Air Masses Transported from Asia to the U.S. West Coast

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Airborne measurements of volatile organic compounds (VOCs) were performed using a proton-transfer-reaction mass spectrometer (PTR-MS) operated onboard a NOAA WP-3 aircraft during the Intercontinental Transport and Chemical Transformation (ITCT) experiment in 2002. Enhancements of acetone (CH₃COCH₃), methanol (CH₃OH), acetonitrile (CH₃CN) and in some cases benzene were observed in air masses that were impacted by outflow from Asia. The enhancement ratios with respect to carbon monoxide are compared to emission factors for fossil fuel combustion and biomass burning, which gives some insight into the sources responsible for the pollution. The observed mixing ratios for acetone, methanol and in particular acetonitrile were generally reduced in the marine boundary layer, suggesting the presence of an ocean uptake sink. The ocean uptake of acetonitrile was found to be particularly efficient in a zone with upwelling water off of the U.S. west coast. Reduced mixing ratios of acetone and methanol were observed in a stratospheric intrusion. This observation gives some information about the lifetime of these VOCs in the stratosphere. Enhanced concentrations of aromatic hydrocarbons were observed in air masses that were impacted by urban sources in California. The ratio between the concentrations of benzene, toluene and higher aromatics indicated the degree of photochemical oxidation. PTR-MS only gives information about the mass of the ions produced by proton-transfer reactions between H₃O⁺ and VOCs in the instrument. The identification of VOCs was confirmed by coupling a gas-chromatographic (GC) column to the instrument and post-flight GC-PTR-MS analyses of canister samples collected during the flights.

A52E-08 1555h

Measurements of Sea Salt Aerosols in the Marine Boundary Layer and Free Troposphere: Vertical Transport and Chemical Transformation

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During the Intercontinental Transport and Chemical Transformation (ITCT) mission (Monterey, CA, spring 2002) nearly 400,000 positive and negative mass spectra of single atmospheric aerosols were acquired using the PALMS (Particle Analysis by Laser Mass Spectrometry) instrument. The primary focus of the mission was to investigate the composition of air masses along the western coast of the United States. Of particular interest to the mission was to study the influence of anthropogenic emissions from Asia on aerosol composition. To accomplish these goals, the WP-3 aircraft, equipped with a suite of instruments including PALMS, covered a large spatial area flying from 0 8000 m altitude covering most of the western coastline from Canada to southern California including flights over the San Francisco and Los Angeles metropolitan areas. The in situ measurements of single particle aerosol mass spectra by PALMS allow for good spatial and vertical

resolution of the aerosol composition. By observing the changes in aerosol composition as a function of altitude, the vertical transport of sea salt aerosols over marine and urban environments is examined. Using measurements of other chemical tracers along with the aerosol composition, the chemical processing of these aerosols during transport both vertically and inland can be discerned. These results add insight into the transport and chemical evolution of sea salt aerosol.

A52E-09 1610h

Measurements of Fine Particle Chemical Composition As Part of the ITCT 2K2 Experiment

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This talk reports on near real-time measurements of fine particle (PM₁) chemical composition as part of the NOAA Intercontinental Transport and Chemical Transformation (ITCT) project conducted in the spring of 2002 (2K2). Airborne measurements from the NOAA WP-3 research aircraft based at Monterey, California and measurements at a ground-based site at Trinidad Head, California will be reported. The approach involved Particle-Into-Liquid Samplers for measurements of a suite of aerosol ions and water-soluble carbon. In this instrument, particles are collected into a flow of purified water for on-line analysis with either a dual channel ion chromatograph or a total carbon analyzer. Fine particle concentrations at Trinidad Head were generally low, averaging around 4 to 5 ug per cubic meter with sulfate as a major component. The regions investigated by the WP-3 included contributions from a variety of sources, including ocean-going shipping vessels, urban areas, such as San Francisco, the Los Angeles basin, and the California Central Valley, and Asian outflow plumes identified by high CO concentrations. Measurements in these various regions, and comparisons of the Asian plumes to our measurements as part of ACE Asia and TRACE P will be discussed.

A52E-10 1625h

A Synthesis of Information on Trans-Pacific Transport of Pollutants: Evidence for Impacts in the Western U.S.

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Trans-Pacific transport of pollutants is now well established. As part of our PHOBEA project (Photochemical Ozone Budget of the Eastern North Pacific Atmosphere) we have identified numerous episodes of pollutants associated with industrial emissions, biomass burning, and mineral dust coming directly from the Eurasian continent. In addition, using the GEOS-Chem global model we can show that even during non-episode periods there are modest impacts associated with global/long-range transported emissions. While most of the observations to date have been in spring, there is some evidence for impacts in other seasons as well.

In this paper we present a summary of evidence for impacts in the Western U.S. associated with this intercontinental transport. For this analysis I will focus on three issues:

- 1) Increasing background surface O₃;
- 2) Increasing wet deposition of nitrate;
- 3) Impact of Eurasian particulate matter on urban areas in the western U.S.

A52E-11 1640h

A comparison of airstream trace gas signatures upwind and downwind of North America

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Recent studies of trace gas export from North America have demonstrated that the transport is closely associated with the airstream flow through mid-latitude cyclones. These cyclones are composed of four primary airstreams: warm conveyor belt (WCB), cold conveyor belt (CCB), dry airstream (DA), and post cold front airstream (PCF). In general the DA advects stratospheric ozone into the mid- and upper troposphere; the WCB exports surface emissions to the upper troposphere and is the most favorable location for photochemical ozone production; the PCF originates to the northwest, and is associated with rapid low altitude transport of surface emissions to the Atlantic; the CCB is generally cloudy and does not show signs of significant photochemical ozone production. The recent Intercontinental Transport and Chemical Transformation (ITCT) aircraft-based measurement campaign has produced chemical measurements in airstreams just west of North America. This new data set provides an excellent opportunity to compare the trace gas signatures of airstreams upwind and downwind of North America. North American trace gas import during ITCT was associated with the remnants of mid-latitude cyclones that had formed over the western North Pacific Ocean. The aged nature of these airstreams and the contrast of emissions ratios between Asia and North America yield different trace gas relationships upwind and downwind of North America. This study examines the relationships between ozone, CO, NO_y and NO in the airstream remnants impacting western North America and compares the results to similar studies conducted for spring 1996 and autumn 1997 on the North American east coast. Conclusions are drawn on the chemical transformation that occurs in air masses traversing North America.

A61A MCC: Hall D Saturday 0830h

Aerosol, Cloud, and Tropospheric Chemistry II Posters

Presiding: C Wang, Massachusetts

Institute of Technology; **S N Pandis, Carnegie Mellon University**

A61A-0040 0830h POSTER

Preliminary Studies on Comparing the Effect of Cloud Chemistry on Tropospheric Oxidants from Modeling a Polydispersed Cloud Drop Population to Modeling a Monodispersed Cloud Drop Population

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Cloud chemistry, that is, both aqueous phase chemistry and the modification of gas-phase chemistry due to separation of reactants, can affect the production

of important tropospheric oxidants. The influence of cloud chemistry on tropospheric oxidants is dependent on the pH of the drops, the liquid water content, and the chemical environment. Because pH varies across the cloud drop spectrum, the degree that cloud chemistry affects tropospheric oxidants is not obvious. For this study, a cloud parcel model that represents cloud drop activation, condensation and coalescence is coupled with a gas-aqueous chemistry module that represents O₃-NO_x-NMHC chemistry appropriate for the remote troposphere. Results of simulations in which cloud drops activate on a polydispersed CCN distribution are compared to results of a simulation in which a single-sized cloud drop population is assumed.

The following hypothesis is tested and discussed. The hypothesis is that the total concentration of species such as O₃, CH₂O, and peroxides will be smaller when a polydispersed cloud drop population is represented than when a single-sized cloud drop population is assumed because previous studies have shown that these oxidants are depleted more or produced less at high pH compared to low pH. This hypothesis assumes that the high pH drops play an important role in the aqueous chemistry as they have previously been shown to do for aqueous sulfur chemistry.

A61A-0041 0830h POSTER

Influence of Biomass Burning on Particle Size Distributions, Hygroscopic and Cloud-nucleating Properties of the Amazon Rain Forest Aerosol

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In the July 2001 transition period between the wet and dry season, aerosol particle size distribution and hygroscopic growth were measured in central Amazonia, as a part of the LBA-CLAIRE experiment. The sub-micron particle number-size distribution were studied with a Differential Mobility Particle Sizer (DMPS) and a Hygroscopic Tandem Differential Mobility Analyzer (H-TDMA) measured the hygroscopic growth at 90% RH for particle dry sizes in the Aitken and accumulation mode size ranges. Three different time periods were selected; a clean time period, a recently yet moderately polluted time period and an aged pollution time period. The pollution was, in both the latter cases, caused by biomass burning.

During the clean time period the observations showed good agreement with the CLAIRE 1998 wet season experiment. During the polluted time periods particle concentrations were substantially increased from about 500 to 2000 part/cc, with a particle number size distribution peak at around 130-140 nm. The biomass burning aerosol frequently showed a bimodal hygroscopic behavior, with a separation into nearly hydrophobic and moderately hygroscopic particles. Besides the increased presence of nearly hydrophobic particles an increase in the hygroscopicity of the moderately hygroscopic particles was observed, compared to the clean case. The numbers of ions in each individual particle were estimated from the H-TDMA data, and together with the DMPS measurements the number of aerosol particles activating to form cloud drops, as a function of water vapor supersaturation, was predicted with a time resolution of 10min. The predicted CCN concentrations were compared with direct measurements performed with a CCN counter, and showed reasonable good agreement.

A61A-0042 0830h POSTER

Modeling of Cloud Droplet Number Concentrations and Their Link to Aerosol and Cloud Chemistry

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The indirect effect of aerosols on clouds is based upon the links between aerosol number concentrations,

activity of particles as cloud condensation nuclei, and cloud drop number concentrations. Modeling and observations have also shown that aqueous-phase chemistry can modify cloud condensation nuclei number concentrations and affect drop concentrations in subsequent cloud cycles. The feedback of chemistry to cloud drop number concentration can be positive or negative, depending upon the initial aerosol size spectrum, chemical environment, and dynamics of each cloud cycle. In this work we focus on the calculation of cloud drop number concentration from a range of specified aerosol initial conditions and examine the sensitivity of predicted drop number concentrations to environmental conditions, model type, and model assumptions.

A61A-0043 0830h POSTER

In Situ Observations of Both Indirect Aerosol Effects

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Cloud microphysics and sub-cloud cloud condensation nuclei (CCN) measurements in polluted, maritime, and transition air masses demonstrated the effects of anthropogenic particles on clouds known as the indirect aerosol effect (IAE). Near-simultaneous cloud droplet measurements showed the higher concentrations and smaller sizes expected for higher CCN concentrations. The associated lower concentrations of large cloud droplets in polluted air indicated that the higher CCN concentrations were responsible for the 1 to 2 orders of magnitude lower drizzle drop concentrations in the polluted clouds. The similarity of the clean and polluted cloud droplet spectra near cloud base, however, suggested that there were no significant differences in giant nuclei concentrations that may have been responsible for greater precipitation in the more maritime clouds. This suppression of warm rain by higher CCN concentrations (2nd IAE) often occurred hundreds of km from anthropogenic sources. These results were similar in: 1) stratocumulus clouds in the eastern Atlantic (ASTEX); 2) small cumulus clouds in eastern Florida (SCMS); and small trade wind cumuli in the Indian Ocean (INDOEX). Comparisons of CCN and cloud droplet spectra showed that cloud supersaturations were lower in the polluted clouds. Thus a smaller percentage of these higher CCN concentrations actually produced cloud droplets. However, the suppression of cloud supersaturation was smaller than previously thought because cloud droplet sizes were so much reduced that it was obvious that many of the polluted cloud droplets were too small to be detected by the forward scattering spectrometer (FSSP). This probably typical undercounting of cloud droplets suggested that the suppression of supersaturations in polluted clouds has probably been overestimated. This means that anthropogenic CCN may have an even greater effect on clouds than previously thought. Moreover, when the missing droplets were accounted for, the relationship between CCN concentrations and cloud droplet concentrations was closer to linear. This suggests that speculations about anomalous effects of some CCN (especially organic material) may be exaggerated.

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Cloud-Sulfate Correlations as a Constraint on Heterogeneous Sulfate Production

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Statistical analysis of observed daily cloud cover (from ISCCP) and sulfate surface concentrations in Europe and North America indicates a significant negative correlation between clouds and sulfate. These anti-correlations are at long time scales (typically 8-64 days) and occur for many months of the 3 years of data. A corresponding analysis of modeled sulfate and cloud cover (from the Goddard Institute for Space Studies GCM) fails to produce significant correlation. However, if we separate heterogeneous (cloud-produced) and gas phase sulfate, there is some significant negative correlation between clouds and gas phase sulfate. If we turn off gas phase sulfate production beneath clouds (as should happen since the oxidant OH is photochemically generated), this negative correlation becomes stronger; however the total sulfate-cloud correlation remains insignificant.

This version ('2-prime', 9-layer) of the GISS model, like most global models, does not have a separate budget for dissolved species. Instead, after each cloud